Spin-dimensionality change induced by Co-doping in the chiral magnet $Fe_{1-x}Co_xSi$

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(Dated: June 12, 2016)

Abstract

Dimensionality is one of the most important parameters in determination of the physical properties. Therefore, tuning of effective dimensionality is of significant importance for modulating the functionality of materials. In this work, we find that the spin-dimensionality can be changed by the Co-doing in $Fe_{1-x}Co_xSi$ system. Investigation of the critical behavior shows that critical exponents for $x = 0.3$ agree with the three-dimensional (3D) Heisenberg model with $\{d : n = 3 : 3\}$ (d is the spatial-dimensionality, and n is the spin-dimensionality). With the increase of Co-content, the critical exponents for $x = 0.5$ fulfill the 3D-XY model with $\{d : n = 3 : 2\}$, while those for $x = 0.6$ approach the 3D-Ising model with $\{d : n = 3 : 1\}$. These results indicate the lowering of the spin-dimensionality with the increase of Co-content in $Fe_{1-x}Co_xSi$. We suggest that the modulation of the spin-dimensionality in $Fe_{1-x}Co_xSi$ should be resulted from the enhancement of the anisotropic magnetic interaction induced by the doping of Co.

PACS numbers: 75.40.-s, 75.40.Cx, 75.40.Gb

Keywords: spin-dimensionality; anisotropic magnetic coupling; crossover phenomenon

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I. INTRODUCTION

The universality class of a phase transition is determined by two factors: the dimensionality of a given system and the symmetry of the order parameter [1]. Therefore, the dimensionality is one of the most important parameters in determination of the physical phenomena. Physical properties are highly sensitive to the variation of the spatial-dimensionality (d) due to confine size effects, such as different characteristics in bulk, film, and one-dimensional wire materials [2, 3]. Many exotic phenomena have been reported to appear in lower spatial-dimensional materials, such as the prominent Peierls phase transition in one-dimensional metallic chain [4]. However, examples in which the effective dimensionality is reduced are quite rare. For a magnetic material, in addition to the spatial dimensionality of the crystal structure, the spin-dimensionality (n) plays an important role in regulation of the magnetic behavior [5]. For example, one-dimensional magnetic coupling with $n = 1$ is manifested as the Ising model, two-dimensional one with $n = 2$ is described with the XY model, and isotropic three-dimensional magnetic coupling with $n = 3$ is depicted with the Heisenberg model [6, 7]. The magnetic coupling can be modulated by the external means, such as magnetic field (H), pressure (P), and chemical doping [8–10]. Theoretically, it has been demonstrated that a crossover phenomenon is prospected to occur in a weakly anisotropic magnetic system [6]. Thus, a tuning of the spin-dimensionality by external means is expected in a weakly anisotropic magnetic system.

In this work, a crossover phenomenon of the spin-dimensionality induced by the chemical doping is found in $Fe_{1-x}Co_xSi$. The B20 compound $Fe_{1-x}Co_xSi$ exhibits a chiral magnetic ordering due to the competition between the ferromagnetic and Dzyaloshinsky-Moriya (DM) interaction [11]. Weak anisotropic magnetic coupling originates from the DM interaction. It has been identified that the DM interaction can be controlled by the doping of Co, and a spin flip occurs at $x \sim 0.65$ [12]. Moreover, a skyrmion phase has been observed at $x \sim 0.5$ [13], and a new type of thermodynamically stable particle-like state has been predicted recently [14]. Through the investigation of the critical behavior of helimagnetic $Fe_{1-x}Co_xSi$ single crystal, we find that the spin-dimensionality changes from three-dimensional magnetic coupling to one-dimensional one with the increase of Co content, which suggests that the effective modulation of spin interaction can be realized by the change of Co-concentration in $Fe_{1-x}Co_xSi$.

II. EXPERIMENT

Single crystal samples of $Fe_{1-x}Co_xSi$ ($x = 0.3$, 0.5 and 0.6) were synthesized by the C-zochralski method [15]. The measurement of magnetization was performed using a Quantum Design vibrating sample magnetometer (SQUID-VSM). The no-overshoot mode was applied to ensure a precise magnetic field. The field was relaxed for two minutes before date collection. To ensure each curve was initially magnetized, the isothermal magnetization was performed after the sample was heated well above T_C for ten minutes, then cooled under zero field to target temperature. The magnetic background was carefully subtracted. The applied magnetic field H_a has been corrected into the internal field as $H = H_a - NM$ (where M is the measured magnetization; N is the demagnetization factor) [16]. The investigation in this work was carried out based on the corrected H.

III. RESULTS AND DISCUSSION

As we know, the spin-dimensionality has significant influence on the magnetic coupling and magnetic behavior. For a ferromagnetic material, the influence of dimensionality on the critical behavior can be manifested through critical exponents. In the vicinity of a second-order phase transition, the spontaneous magnetization M_S and initial susceptibility χ_0 are correlated with the critical exponents as [17, 18]:

$$M_S(T) = M_0(-\varepsilon)^{\beta}, \varepsilon < 0, T < T_C \tag{1}$$

$$\chi_0^{-1}(T) = (h_0/M_0)\varepsilon^{\gamma}, \varepsilon > 0, T > T_C \tag{2}$$

$$M = DH^{1/\delta}, \varepsilon = 0, T = T_C \tag{3}$$

where $\varepsilon = (T - T_C)/T_C$ is the reduced temperature; M_0/h_0 and D are critical amplitudes. The parameters β (associated with M_S), γ (associated with χ_0) and δ (associated with T_C) are the critical exponents. Generally, these critical exponents should follow the Arrott-Noakes equation of state in the asymptotic critical region [19]:

$$(H/M)^{1/\gamma} = (T - T_C)/T_C + (M/M_1)^{1/\beta} \tag{4}$$

Therefore, the spin coupling can be revealed by investigating the critical behavior. The critical temperature T_C can be roughly determined from the temperature dependence of

magnetization [$M(T)$]. Figure 1 (a) depicts $M(T)$ curves for $Fe_{1-x}Co_xSi$ (x = 0.3, 0.5 and 0.6) under zero-field-cooling (ZFC) and field-cooling (FC) with an applied magnetic field H = 10 Oe. With temperature increasing, all three samples undergo helimagnetic-paramagnetic (HM-PM) phase transitions. The T_C are determined as 52 K, 46 K, and 28 K for x = 0.3, 0.5, and 0.6 respectively. Figure 1 (b) plots the isothermal magnetization $M(H)$ at 4 K, which shows that all three samples exhibit magnetic ordering behaviors. The inset of Fig. 1 (b) plots the magnification of $M(H)$ in lower field region, which indicates that almost no coercive force.

According to Eqs. (1) and (2), the critical exponents β and γ can be obtained by fitting the $M_S(T)$ and $\chi_0^{-1}(T)$ curves based on the modified Arrott plot of $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$. Therefore, the initial isothermal $M(H)$ curves around T_C are measured to obtain the critical exponents, as shown in Figs. 2 (a), (b) and (c). Previous investigation has suggested that $Fe_{1-x}Co_xSi$ with $x = 0.2$ could be described within the 3D-Heisenberg model [20]. Therefore, the modified Arrott plot of $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ is constructed within the framework of the 3D-Heisenberg model with β = 0.365 and γ = 1.386, as depicted in Figs. 2 (d), (e) and (f). The $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ curves exhibit straight lines with positive slopes in the high field region, suggesting a second-order transition [21, 22]. The $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ curve at T_C passes the origin. The linear extrapolation from the high field region to the intercepts with $M^{1/\beta}$ and $(H/M)^{1/\gamma}$ axes yields reliable values of $M_S(T, 0)$ and $\chi_0^{-1}(T, 0)$, which are plotted as a function of temperature in Fig. 3 (a), (b) and (c). Subsequently, according to Eqs. (1) and (2), we obtain that $\beta = 0.387(3)$ and $\gamma = 1.382(8)$ for $x = 0.3$, $\beta = 0.357(6)$ and $\gamma = 1.293(3)$ for $x = 0.5$, $\beta = 0.331(2)$ and $\gamma = 1.230(4)$ for $x = 0.6$. The obtained exponents are listed in Table I.

The critical exponent δ can be derived by the isothermal magnetization $M(H)$ at T_C following Eq. (3). Figures 3 (d), (e) and (f) show the $M(H)$ at T_C, and the insets give those on $\log - \log$ scale. We obtain that $\delta = 4.678(1)$ for $x = 0.3$, $\delta = 4.574(2)$ for $x = 0.5$, and $\delta = 4.452(1)$ for $x = 0.6$ (see Table I). According to statistical theory, these critical exponents should fulfill the Widom scaling relation [23]:

$$\delta = 1 + \frac{\gamma}{\beta} \tag{5}$$

As a result, $\delta = 4.571(8)$ for $x = 0.3$, $\delta = 4.622(6)$ for $x = 0.5$, and $\delta = 4.716(4)$ for $x = 0.6$ are calculated following the Widom scaling relation. These calculated results agree well with

those yielded from the experimental critical isotherm analysis. The self-consistency of these critical exponents demonstrates that they are reliable and unambiguous.

As predicted by the scaling equation, the $M-T-H$ curves can be scaled into a universality class using these critical exponents. According to scaling equation, in the asymptotic critical region the magnetic equation can be written as [18]:

$$M(H,\varepsilon)=\varepsilon^{\beta}f_{\pm}(H/\varepsilon^{\beta+\gamma}) \tag{6}$$

where $f_{\pm}$ are regular functions with f_{+} for $T>T_C$ while f_{-} for $T<T_C$. Defining the renormalized magnetization as $m\equiv\varepsilon^{-\beta}M(H,\varepsilon)$ and renormalized field as $h\equiv H\varepsilon^{-(\beta+\gamma)}$, the scaling equation indicates that m vs. h forms two universal curves for $T>T_C$ and $T<T_C$, respectively [10, 24]. Based on the scaling equation [$m=f_{\pm}(h)$], the isothermal magnetization around the critical temperatures for $Fe_{1-x}Co_xSi$ are replotted in Fig. 4 (a), (b) and (c) on $\log-\log$ scale. All $M-T-H$ curves collapse onto two universal branches, which further confirms the reliability of the obtained critical exponents.

The obtained critical exponents of $Fe_{1-x}Co_xSi$ ($x=0.2$, 0.3, 0.5, 0.6), as well as those of different theoretical models and related materials, are listed in Table I for comparison [25–28]. It can be seen that the critical exponents of $Fe_{0.7}Co_{0.3}Si$, which approach those of the $Fe_{0.8}Co_{0.2}Si$, are close to the 3D-Heisenberg model. With the increase of Co-content, the critical exponents of $Fe_{0.5}Co_{0.5}Si$ agree with those of the 3D-XY model. The critical exponents of $Fe_{0.4}Co_{0.6}Si$ approach the prediction of the 3D-Ising model. Theoretical expectation indicates that the variational theoretical models correspond to different spin-dimensionality. For the 3D-Heisenberg model, the spin coupling is isotropic with $\{d:n=3:3\}$ [29], while it is $\{d:n=3:2\}$ for the 3D-XY model and $\{d:n=3:1\}$ for the 3D-Ising model [29]. That is to say, with the doping of Co, the spin-dimensionality changes from $n=3$ to $n=1$. For the spin interaction, there are $\overrightarrow{S}=S(S_x,S_y,S_z)$ for $n=3$, $\overrightarrow{S}=S(S_x,S_y)$ for $n=2$ and $\overrightarrow{S}=S(S_z)$ for $n=1$, as shown in Fig. 5 (a), (b), and (c) [6]. M. E. Fisher has predicted the crossover of the spin-dimensionality by the renormalization group theory [6]. It has been suggested that weakly anisotropic magnetic interaction would induce the crossover of spin-dimensionality from $n=3$ to $n=1$ [6]. As we know, the existence of DM interaction in $Fe_{1-x}Co_xSi$ results in weakly anisotropic magnetic interaction. Moreover, it has been demonstrated that the DM interaction can be tuned by Co-doping [12]. Therefore, the change of spin-dimensionality can be realized by the change of Co-content through the

modulation of the DM interaction.

The spin-dimensionality n is manifested through γ as [5, 30]:

$$\gamma(n) = 2\frac{n+4}{n+7} \tag{7}$$

Theoretically, it is obtained that $\gamma(3) = 1.4$, $\gamma(2) = 1.33$, and $\gamma(1) = 1.25$. One can see that for higher n-values the formula gives lower γ-values. Therefore, the decrease of γ with increasing x in $Fe_{1-x}Co_xSi$ indicates the lowering of the spin-dimensionality. For a homogeneous magnet, the universality class of the magnetic phase transition depends on the exchange distance $J(r)$. A renormalization group theory study gives that $J(r)$ depends on the spatial distance r as [31, 32]:

$$J(r) \approx r^{-(d+\sigma)} \tag{8}$$

were σ is a positive constant. Meanwhile, σ is determined by γ as [16, 29, 31]:

$$\gamma = 1 + \frac{4}{d}\frac{n+2}{n+8}\Delta\sigma + \frac{8(n+2)(n-4)}{d^2(n+8)^2} \times \left[1 + \frac{2G(\frac{d}{2})(7n+20)}{(n-4)(n+8)}\right]\Delta\sigma^2 \tag{9}$$

where $\Delta\sigma = (\sigma - \frac{d}{2})$ and $G(\frac{d}{2}) = 3 - \frac{1}{4}(\frac{d}{2})^2$. When $\sigma \geq 2$, the Heisenberg model ($\beta = 0.365$, $\gamma = 1.386$ and $\delta = 4.8$) is valid, revealing that $J(r)$ decreases faster than $r^{-(d+2)}$. When $\sigma \leq 3/2$, conditions for the mean-field model ($\beta = 0.5$, $\gamma = 1.0$ and $\delta = 3.0$) are satisfied, expecting that $J(r)$ decreases slower than $r^{-(d+1.5)}$. From Eq. 9, it is obtained that σ=1.9324(2) for $x = 0.2$, σ=1.9341(5) for $x = 0.3$, σ=1.8965(7) for $x = 0.5$, and σ=1.8879(2) for $x = 0.6$. Finally, for $x = 0.2$ and $x = 0.3$ with $\{d : n = 3 : 3\}$, we yield $J(r) \approx r^{-4.93}$. For $x = 0.5$ with $\{d : n = 3 : 2\}$, we obtain $J(r) \approx r^{-4.90}$. For $x = 0.6$ with $\{d : n = 3 : 1\}$, we deduce $J(r) \approx r^{-4.89}$. The decrease of σ indicates that the spatial decay distance of magnetic coupling increases with the increase of the Co-content, however, still belongs to a short-range magnetic coupling. Moreover, $\nu = \gamma/\sigma$ ($\xi = \xi_0|(T - T_C)/T_C|^{-\nu}$) and $\alpha = 2 - \nu d$ ($C_p = A^{\pm}\varepsilon^{-\alpha}$) can also be obtained, as listed in Table I. The change of α from negative to positive confirms the lowering of the spin-dimensionality of the magnetic interaction [33].

The phase diagram of the critical exponents as a function of Co-content x for $Fe_{1-x}Co_xSi$ is shown in Fig. 5 (d). From the phase diagram, it is concluded that spin interaction belongs to the 3D-Heisenberg model with $n = 3$ when $x \lesssim 0.45$. In the range of $0.45 \lesssim x \lesssim 0.55$, the spin coupling is close to the 3D-XY model with $n = 2$. When $x \gtrsim 0.55$, it approaches

the 3D-Ising model with $n = 1$. The results suggest that the doping of Co enhances the anisotropic magnetic interaction in $Fe_{1-x}Co_xSi$. It has been demonstrated that in the B20 compounds, the magnetism correlates closely with the structure, and the DM interaction in $Fe_{1-x}Co_xSi$ can be effectively controlled by the Co composition [12, 34]. Thus, the change of spin-dimensionality induced by the doping of Co should be caused by the modulation of the DM interaction. It is noticed that the skyrmion state emerges at $x \sim 0.5$ just lying in the region of $\{d : n = 3 : 2\}$, which implies that two-dimensional magnetic coupling may be in favor of the formation of skyrmion phase.

IV. CONCLUSION

In summary, the critical behaviors of chiral magnets $Fe_{1-x}Co_xSi$ are investigated by the means of bulk dc-magnetization. We found that the critical exponents for $x = 0.3$ are close to the 3D-Heisenberg model with $\{d : n = 3 : 3\}$. With the increase of Co-content, the critical exponents for $x = 0.5$ fulfill the 3D-XY model with $\{d : n = 3 : 2\}$, while those for $x = 0.6$ satisfy the 3D-Ising model with $\{d : n = 3 : 1\}$. These results indicate that the spin-dimensionality n changes from three-dimensional to one-dimensional with the increase of Co content. We suggest that the modulation of the spin-dimensionality should be resulted from the enhancement of anisotropic magnetic interaction induced by the doping of Co in $Fe_{1-x}Co_xSi$.

V. ACKNOWLEDGEMENTS

This work was supported by the State Key Project of Fundamental Research of China through Grant No. 2011CBA00111, the National Natural Science Foundation of China (Grant Nos. 11574322, U1332140, 11004196, U1232142,, 11474290, 11104281, and 11204288), the Foundation for Users with Potential of Hefei Science Center (CAS) through Grant No. 2015HSC-UP001.

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TABLE I: Comparison of critical exponents of $Fe_{1-x}Co_xSi$ with different theoretical models and related materials (MAP = modified Arrott plot; Hall = Hall effect; AC = ac susceptibility; SC = single crystal; PC = polycrystal).

Composition	technique	Ref.	T_C(K)	α	β	γ	δ	σ	ν
$Fe_{0.8}Co_{0.2}Si^{PC}$	Hall	[20]	36.0	-0.1424(1)	0.371(1)	1.38(2)	4.78(1)	1.9324(2)	0.7141(3)
$Fe_{0.7}Co_{0.3}Si^{SC}$	MAP	This work	48.2(1)	-0.1436(3)	0.387(3)	1.382(8)	4.678(1)	1.9341(5)	0.7145(4)
$Fe_{0.5}Co_{0.5}Si^{SC}$	MAP	This work	43.1(1)	-0.0453(4)	0.357(6)	1.293(3)	4.574(2)	1.8965(7)	0.6817(8)
$Fe_{0.4}Co_{0.6}Si^{SC}$	MAP	This work	25.8(2)	0.0454(4)	0.331(2)	1.230(4)	4.452(1)	1.8879(2)	0.6515(2)
3D-Heisenberg	theory	[7]	-	-0.115(9)	0.365	1.386	4.80	-	-
3D-XY	theory	[7]	-	-0.007(6)	0.346	1.316	4.81	-	-
3D-Ising	theory	[7]	-	0.110(5)	0.325	1.241	4.82	-	-
Tricritical mean-field	theory	[25]	-	-	0.25	1.0	5.0	-	-
Mean-field	theory	[7]	-	-	0.5	1.0	3.0	-	-
$Cu_2OSeO_3^{SC}$	AC	[26]	58.3	-	0.37(1)	1.44(4)	4.9(1)	-	-
$MnSi^{SC}$	MAP	[27]	30.5	-	0.242(6)	0.915(3)	4.734(6)	1.329(8)	0.688(9)
$FeGe^{PC}$	MAP	[28]	283	-	0.336(4)	1.352(3)	5.267(1)	1.908(7)	0.709(8)

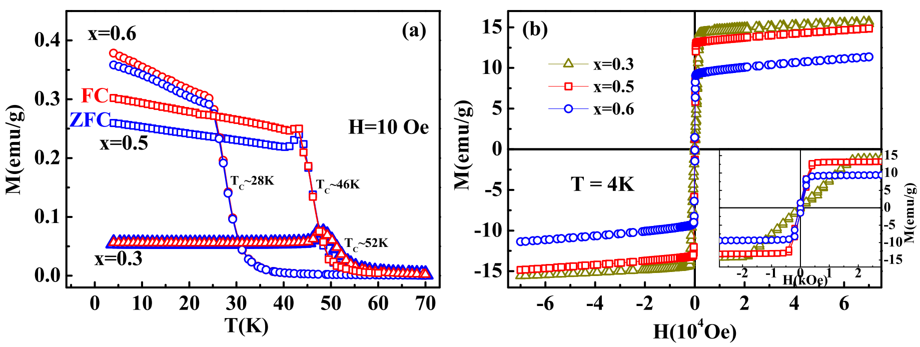

FIG. 1: (Color online) (a) The temperature dependence of magnetization [$M(T)$] under ZFC and FC for $Fe_{1-x}Co_xSi$ (x = 0.3, 0.5, and 0.6); (b) the isothermal magnetization [$M(H)$] at 4 K (the inset gives that in low field region).

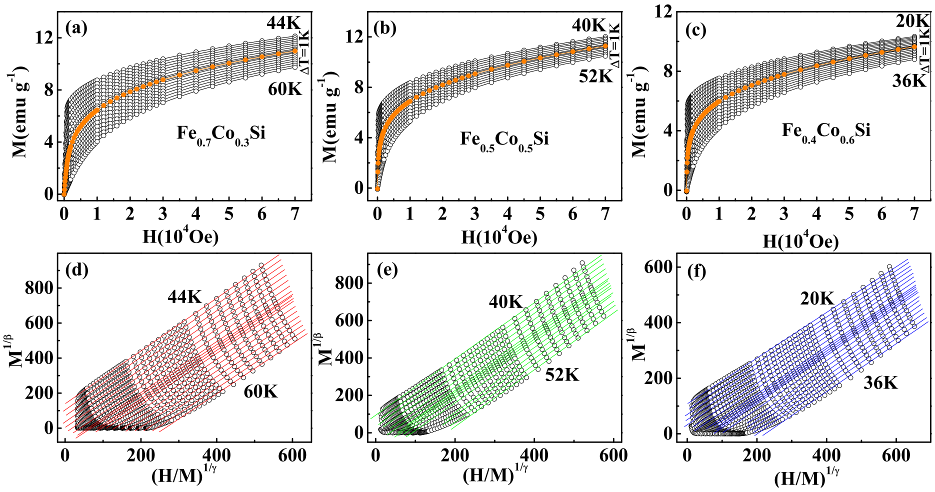

FIG. 2: (Color online) (a), (b), and (c): The initial magnetization around T_C for $Fe_{1-x}Co_xSi$ ($x =$ 0.3, 0.5, and 0.6); (c), (d), and (e): modified Arrott plots of $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ with $\beta = 0.365$ and $\gamma = 1.386$ (the solid lines are fitted).

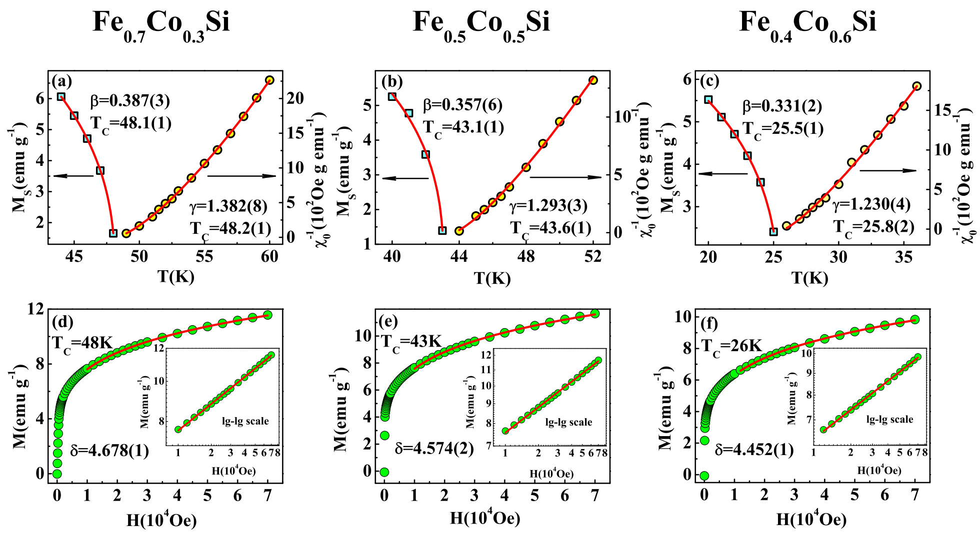

FIG. 3: (Color online) (a), (b), and (c): The temperature dependence of M_S and $\chi_0{}^{-1}$ for $Fe_{1-x}Co_xSi$ (x = 0.3, 0.5, and 0.6); (d), (e), and (f): the isothermal $M(H)$ at T_C with the $\log-\log$ scale in the inset (the solid curves are fitted).

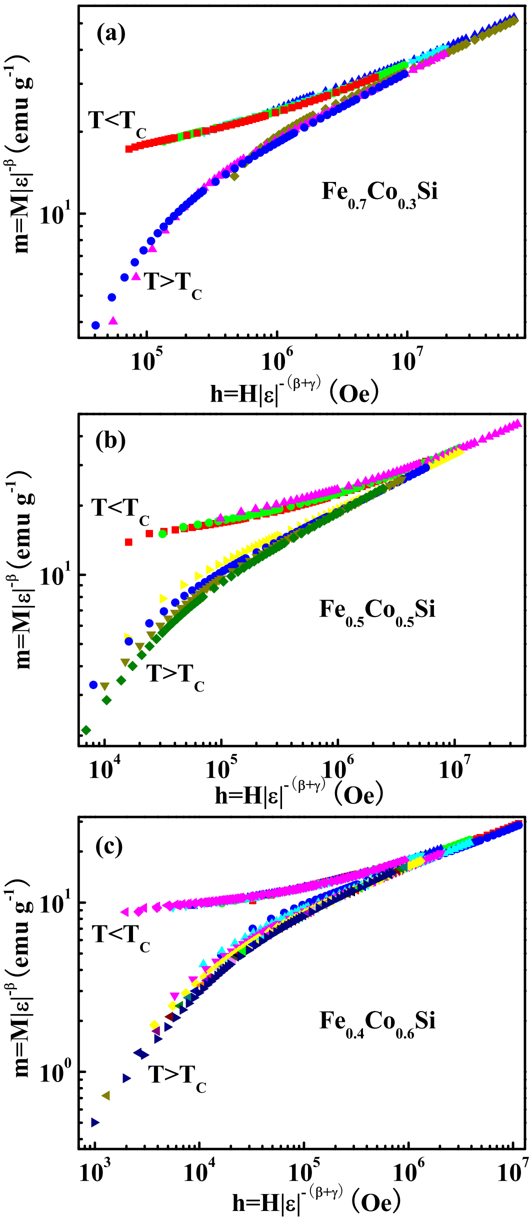

FIG. 4: (Color online) Scaling plots of renormalized magnetization m vs. renormalized field h around T_C for $Fe_{1-x}Co_xSi$ (x = 0.3, 0.5, and 0.6)

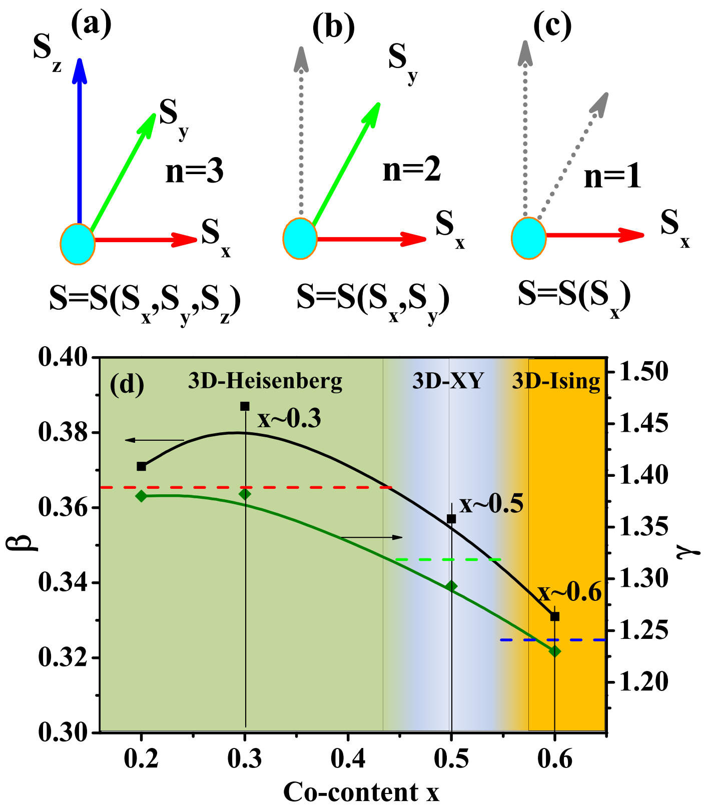

FIG. 5: (Color online) The sketched illustration for the spin interaction with (a) $n = 3$, (b) $n = 2$, and (c) $n = 1$; (d) the change of β and γ as a function of x (the dashed lines mark the theoretical value for the 3D-Heisenberg (red), 3D-XY (green), and 3D-Ising (blue) models).